PATENT SPECIFICATION (11) 1 513 657

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(21) Application No. 260/77 (22) Filed 5 Jan. 1977

(31) Convention Application No. 51/089682 (32) Filed 29 Jul. 1976 in

(33) Japan (JP)

(44) Complete Specification Published 7 Jun. 1978

(51) INT. CL.[2] C08L 23/02
C08K 3/34 5/02 5/06

(52) Index at Acceptance
C3K 111 120 123 129 221 222 292 EA

(54) FLAME RETARDING RESIN COMPOSITION

(71) We, IDEMITSU KOSAN COMPANY LIMITED, a corporation organized and existing under the laws of Japan of No. 1-1, 3-chome, Marunouchi, Chiyoda-ku, Tokyo, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

The present invention relates to a flame-retarding resinous composition.

It has been generally known that the flame retarding property of thermoplastic resin is obtained by adding a flame retarder and/or a flame retarding assistant such as an organic halide, an organophosphorus compound, or antimony oxide, to the aforesaid resin. However, examination of individual thermoplastic resins has shown that the attained degree of flame retardation is different for each resin due to the difference in its properties, and addition of a flame retarder can have a deleterious effect upon the properties of a resin thus making it unsuitable for the use to which the resin could otherwise be put. Of the various resins, it is extremely difficult, especially, with polyolefins to make them flame resistant without adversely effecting their advantageous properties.

With regard to improving the flame retardation of polyolefins, it is already known to be possible in some degree by adding the aforesaid flame retarder thereto.

It is also general knowledge that addition of talc to a resin at least minimizes the tendency at combustion of a polymer to form droplets which can also be a fire risk (Japanese Patent Publication Gazettes 32288/1972 and 6105/1972, 345/1974). The electrical industries demand thin moldings of polyolefin because of its good electrical properties. However, fire resistance is reduced and formation of droplets of resin at combustion occurs more easily as the moldings are made thinner. However, the aforesaid techniques of reducing the combustibility of polyolefin can not provide a flame retarding property and prevent droplet formation for thin moldings of polyolefin.

The present invention provides a composition comprising 30 to 80 percent by weight of polypropylene, 5 to 25 percent by weight of polyethylene, 10 to 50 percent by weight of at least one kind of inorganic filler selected from powdered talc, kaolinite, sericite, silica and diatomaceous earth, and a flame retarder, the percentage amount of each component being based on the total amount of polypropylene, polyethylene and inorganic filler present in the composition.

Polypropylene constitutes the basis for the composition of the present invention and determines the fundamental physical properties of the composition. As polypropylene, copolymers of propylene with some amount of other olefins as well as homopolymers of propylene can be used. Polypropylene with 0.5 to 15.0 grams per 10 minutes of melt index (temperature 230°C, load 2.16 kilograms) is preferred. As stated above, the polypropylene should be present in a composition according to the invention in amounts of 30 to 80% by weight based on the total amount of polypropylene, polyethylene and inorganic filler present in the composition.

The polyethylene component used in a composition according to the present invention enhances the impact strength of the composition and at the same time improves resistance to combustion and shape-retention at combustion. In particular it acts very effectively to prevent droplet formation at combustion. Therefore the amount of flame retarder and flame retarding assistant can be reduced by adding polyethylene to the composition. As the polyethylene component, any low density-, middle density and high density-polyethylene can be used, but especially polyethylene with 0.01 to 2.0 grams per 10 minutes of melt index (temperature

190°C, load 2.16 kilograms) is preferred. The polyethylene component should, as stated above, be present in amounts of 5 to 25% by weight based on the total amount of polypropylene, polyethylene and inorganic filler present in the composition. Amounts of less than 5% by weight cannot prevent droplet formation of obtained moldings when these are combusted. On the other hand, more than 25% by weight is to be avoided because it cannot fully prevent droplet formation at combustion of the obtained moldings and additionally lowers molding ability, thereby affecting the appearance of the moldings, lowering the heat distortion temperature and the rigidity of the moldings to a significant extent.

As the inorganic filler component of a composition according to the present invention, powdered talc, kaolinite, sericite, silica and diatomaceous earth can be used singly or in combination. The average particle size of the filler is preferred to be less than 3μ. In a composition according to the present invention, the inorganic filler serves to improve flame resistance and rigidity of the obtained moldings, to raise the heat distortion temperature, to improve shape-retention at combustion and to prevent droplet formation effectively. The inorganic filler shows an action similar to that of a flame retarder. Therefore the amount of flame retarder can be reduced by the addition of such filler. The amount of inorganic filler in a composition according to the invention should, as stated above, be 10 to 50% by weight based on the total amount of polypropylene, polyethylene and inorganic filler present in the composition. Any improvement in shape retention at combustion is insignificant with amounts of filler less than 10% by weight, whilst on the other hand amounts exceeding 50% by weight induce unfavourable properties such as the increase of density in the obtained molding and the lowering of impact strength and molding ability.

In addition, a composition of the present invention contains a flame retarder in order to improve the flame resistance of the composition and molding made therefrom. Although there is no particular limitation upon the kind of flame retarder which can be used, organic halides, such as decabromodiphenyl ether, dodecachlorododecahydrodimethanobenzocyclo octene, are preferred. Chlorinated paraffin lowers rigidity, the heat distortion temperature and impact strength and consequently promotes droplet formation at combustion. It also induces unfavourable effects such as the lowering of thermal stability. Organic halides such as tetrabromobutane or hexabromobenzene have limited application due mainly to their volatility. The flame retarder may be added in amounts corresponding to the required degree of flame retardation and there is no particular limitation, but usually the range of 5 to 35% by weight based on the 100 parts by weight of the total amount polypropylene, polyethylene and inorganic filler present in a composition according to the invention is preferred. Amounts of less than 5% by weight cannot provide sufficient flame retardation. Amounts of more than 35% by weight reduce the impact strength significantly and increase the cost.

In a composition according to the present invention, antimony compounds, such as antimony trioxide, antimony trichloride and antimony trisulfide, or boron compounds such as zinc borate and borax, can be added to assist flame retardation as required. The amount of additive can be properly decided in accordance with the required degree of flame retardation, but it is usually preferred to be 1/4 to 1/2 of the amount of flame retarder proper. The action of the additive is synergetic with that of the flame retarder. Especially in the field of electrical industries, where the products are usually thin moldings, there is a requirement for a high degree of flame retardation and for materials with high mechanical strength and the use of such flame retarding additive is useful. The addition of flame retarding additive can reduce the amount of flame retarder proper which would otherwise be required.

To a composition according to the present invention, it may of course be desirable to add also coloring agent such as a dyestuff or pigment, nucleation agents, lubricants, plasticizers, heat-resistance stabilizers, ultraviolet absorbers, mold releasing agents, cross-linking agents, and foaming agents, according to the use.

The process for molding a composition according to the present invention is not particularly specified. Usually each aforementioned component is put into a Banbury mixer, or other type of mixer at specific ratio, and mixed while heating. The mixture is formed into a sheet and then made into a pellet with a pelletizer after it has cooled and solidified. According to another process, each ingredient is continuously mixed, and then subjected to strand extrusion. A pellet is obtained by pelletizing the strand with a strand-cutter after it has cooled and solidified.

Moldings subsequently obtained possess a high degree of flame retardation and are excellent in shape-retention at combustion, thus resulting in diminution of droplet formation. In these moldings, sufficient flame retardation can be given even when flame retarder is present in small amounts, due to the presence of polyethylene and inorganic filler.

A composition according to the present invention can be effectively utilized as electrical insulation material, for casings for electrical and electronic components, and for components and accessories in motor vehicles.

Examples and Comparative Examples

A number of experiments were performed

upon examples of compositions according to the invention and of comparitive compositions. In each case, predetermined amounts of each of polypropylene, polyethylene, inorganic filler, flame retarder and antimony trioxide as required were put into a Banbury mixer heated at 150°C (casing temperature) and mixed. Then the mixed composition was made into a sheet and then pelletized.

The thus-obtained pellet was subjected to injection molding at the highest cylinder temperature of 210°C and mold temperature of 50°C to give a piece A for combustion test of dimensions 5 inches x 1/2 inch x 1/16 inch and a piece B of dimensions 5 inches x 1/2 inch x 1/32 inch. The flame retarding properties were tested for each piece. The flame retarding test was performed according to. UL subject 94 (Underwriters Laboratory Incorporation) "Combustion test on plastic materials for the parts of machinery and appliances" to check the classification of burning quality and presence or absence of droplet formation and the ignition of underlaid cotton due to droplet formation. The results are shown in Tables 1 and 2.

WHAT WE CLAIM IS:-

1. A composition comprising 30 to 80 percent by weight of polypropylene, 5 to 25 percent by weight of polyethylene, 10 to 50 percent by weight of at least one kind of inorganic filler selected from powdered talc, kaolinite, sericite, silica and diatomaceous earth, and a flame retarder, the percentage amount of each component being based on the total amount of polypropylene, polyethylene and inorganic filler present in the composition.

2. A composition according to Claim 1 wherein the flame retarder is an organic halide.

3. A composition according to Claim 2 wherein the organic halide is selected from decabromodiphenyl ether, dodecachloro-dodecahydrodimethanobenzo cyclo octene and mixtures thereof.

4. A composition according to anyone of the preceding claims wherein the amount of flame retarder is from 5 to 35 parts by weight based on 100 parts by weight of the total amount of polypropylene, polyethylene and inorganic filler present in the composition.

5. A composition according to anyone of the preceding claims wherein polyethylene is polyethylene with 0.01 to 2.0 grams per 10 minutes of melt index (temperature: 190°C, load: 2.16 kilograms).

6. A composition according to claim 1 substantially as hereinbefore described.

7. A composition according to claim 1 substantially as hereinbefore described with reference to any one of Examples 1 to 14.

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Printed for Her Majesty's Stationery Office, by Croydon Printing Company Limited, Croydon, Surrey, 1978.
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.

TABLE 1

No.	Amount of Component (parts by weight) *1 Poly-propylene	*2 Poly-ethylene	*3 Inorganic filler	*4 Flame retarder	Antimony trioxide
Comparative Example 1	70	0	30	30	10
Example 1	60	10	"	"	"
"	50	20	"	"	"
Comparative Example 2	40	30	"	"	"
" 3	20	50	"	"	"
" 4	0	70	"	"	"
" 5	80	0	20	40	13
Example 3	70	10	"	"	"
" 4	60	20	"	"	"
Comparative Example 6	40	40	"	"	"
" 7	0	80	"	"	"
" 8	60	0	40	12	4
Example 5	50	10*5	"	"	"
" 6	40	20*5	"	"	"
Comparative Example 9	20	40*5	"	"	"
" 10	0	60*5	"	"	"
Example 7	60	5/5*6	30	27	9
" 8	55	7.5/7.5*6	"	"	"
" 9	60	10	"	20/10*7	10
" 10	60	10	"	10/20*7	"

*1 Density: 0.91 grams per cubic centimeter, Melt index: 9 grams per 10 minutes (Temperature 230°C, Load 2.16 kilograms)

*2 Density: 0.955 grams per cubic centimeter, Melt index: 0.05 grams per 10 minutes (Temperature 190°C, Load 2.16 kilograms)

*3 Talc (Average particle size 2.8u)

*4 Decabromodiphenyl ether (Average particle size 2.8u, Bromine content: 83.48 per cent)

*5 Density: 0.964 grams per cubic centimeter, Melt index: 0.4 grams per 10 minutes (Temperature 190°C, Load 2.16 kilograms)

*6 Density: 0.919 grams per cubic centimeter, Melt index: 1.0 grams per 10 minutes (Temperature 190°C, Load 2.16 kilograms)

*7 Dodecachlorododecahydrodimethanobenzo cyclooctene (Average particle size: 3.0u, Chlorine content: 65 per cent)

TABLE 1 continued

No.	Piece A		Piece B		
	Classification of Burning Quality	Fusion Dropping	Classification of Burning Quality	Fusion Drooping	Ignition of Cotton underlaid
Comparative Example 1	-	-	V 0	Yes	No
Example 1	-	-	"	No	"
" 2	-	-	"	"	"
Comparative Example 2	-	-	V 2	Yes	Yes
" 3	-	-	"	"	"
" 4	-	-	"	"	"
" 5	-	-	"	"	"
Example 3	-	1	V 0	No	No
" 4	-	-	"	"	"
Comparative Example 6	-	-	V 2	Yes	Yes
" 7	-	-	"	"	"
" 8	V 0	Yes	-	-	No
Example 5	"	No	-	-	"
" 6	"	"	-	-	"
Comparative Example 9	V- 2	Yes	-	-	Yes
" 10	"	"	-	-	"
Example 7	-	-	V 0	No	No
" 8	-	-	"	"	"
" 9	-	-	"	"	"
" 10	-	-	V 1	"	"

TABLE 2

No.	Component *1 Amount of Polypropylene	*2 Amount of Polyethylene	Inorganic filler Kind	Average particle size (u)	Amount (parts by weight)	*3 Flame retarder (parts by weight)
Example 11	60	10	Kaolinite	3.0	30	30
" 12	"	"	Sericite	2.6	"	"
" 13	"	"	Silica	1.8	"	"
" 14	"	"	Diatomacetous earth	4.1	"	"
Comparative Example 11	"	"	Heavy calcium carbonate	3.0	"	"
" 12	"	"	Calcium sulfite	7.0	"	"
" 13	"	"	Calcined alumina	5.3	"	"
" 14*4	"	"	Aluminum hydroxide *5	5.0	"	"
" 15*4	"	"	Magnesium hydroxide	7.2	"	"
" 16	"	"	Calcium hydroxide	6.3	"	"
" 17	"	"	Calcium silitate	7.9	"	"
" 18*4	"	"	Basic Magnesium carbonate	0.5	"	"

*1 Density: 0.91 grams per cubic centimeter, Melt index: 9 grams per 10 minutes (Temperature 230°C, Load 2.16 kilograms)

*2 Density: 0.955 grams per cubic centimeter, Melt index: 0.05 grams per 10 minutes (Temperature 190°C, Load 2.16 kilograms)

*3 Decafbromodiphenyl ether (Average particle size: 2.8u, Bromine content: 83.48 per cent)

*4 No fusion dropping and good in flame retardation, but appearance of the moldings is inferior due to foaming at the time of molding.

*5 Hydrated alumina having dibbsite crystalline structure.

TABLE 2 continued

Component continued — Piece B

No.	Antimony trioxide (parts by weight)	Classification of Burning Quality	Fusion Dropping	Ignition of Cotton Underlaid	Appearance of Moldings
Example 11	10	V 0	No	No	Good
" 12	"	"	"	"	"
" 13	"	"	"	"	"
" 14	"	"	"	"	"
Comparative Example 11	"	V 2	Yes	Yes	"
" 12	"	"	"	"	"
" 13	"	"	"	"	"
" 14*4	"	V 0	No	No	Inferior (foamed)
" 15*4	"	"	"	"	"
" 16	"	"	Yes	"	Inferior (coarse)
" 17	"	V 2	"	Yes	Good
" 18*4	"	V 0	No	No	Inferior (foamed)